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Chris Dayton
Kaiser-Hill, LLC
Rocky Fats Environmental Technology Site
Golden, CO 80402

16 December 1997

Dear Chris:

Enclosed is a copy of our final report, 'Actinide Migration Studies at the Rocky Flats Environmental Technology Site'. The final report has been modified from the review draft of 19 October 1997 as follows:

1. Review comments of the 19 October 1997 draft from Kirk Nordstron, Davie Clark and Dave Janecky have been incorporated into the final report.

2. Additional data have been included:
   a) Completion of the analyses of the 'residual' fraction. Note: duplicates 1D and 2D could not be quantified because of poor tracer recovery.
   b) K_d kinetics data for $^{239,240}Pu$ desorption; a discussion of U(VI) sorption kinetics.

3. A re-evaluation of the homogenate data with the consideration of a correction factor for the core-depth penetration.

4. Figure 3 (% organic carbon *v.* log Pu_T) was deleted.

5. An Executive Summary has been added.

You will need to replace Figure 1 with an official version from RMRS.

Sincerely,

Bruce D. Honeyman
Associate Professor

cc: John Hopkins, RMRS

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ACTINIDE MIGRATION STUDIES AT THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE

P.I.s: Bruce D. Honeyman, Colorado School of Mines, Golden, CO 80401
Peter H. Santschi, Texas A&M University, Galveston, TX 77551

Contractor: Rocky Mountain Remediation Services
Contract: May 1, 1997 to September 30, 1997; $60,000 unburdened.
Date: 15 December 1997

Final Report

Executive summary:

1. Selective leaching analysis of soil isolates taken from a region trending SE of the 903 Pad indicates: a) from 0.04 to 0.09 % of the soil-associated $^{239,240}Pu$ is 'exchangeable; b) addition of reducing agents 'solubilized' 0.1 to 5% of soil-associated Pu, suggesting that a relatively small percentage of total soil $^{239,240}Pu$ could be released from the soil matrix during prolonged periods of soil anoxia; however such a fraction if solubilized could significantly elevate the aqueous concentration of Pu and explain some of the observed surface water quality exceedances; c) the predominant phase classes for Pu are the 'organic' and 'residual' fractions. The significance of the 'organic' fraction with respect to Pu speciation is not yet clear.
2. Distribution coefficients for $^{239,240}Pu$ interaction with soil isolates under oxic conditions range from ca. 1×10^4 L kg^{-1} to 1.2×10^5 L kg^{-1}. K_d values are dependent on time of contact of solution with soil particles as well as solution composition. The data set described here provides a likely upper range of K_d values.
3. Solid phase extraction studies indicate that $^{239,240}Pu$ has a limited solubility under oxidizing conditions and suggests that the primary transport process under oxidizing conditions is through mechanical erosion.
4. Yearly fluxes of $^{239,240}Pu$ to the RFETS ponds evaluated range from 0.02 to 1.7 mCi y^{-1} depending on the pond and method of Pu inventory determination.
5. K_d values for $^{238}U(VI)$ interaction with solar pond core isolates range under oxidizing conditions range from ca. 30 to 180 L kg^{-1}. Results indicate that U(VI) may be subject to groundwater transport.
6. Additional studies need to be initiated to determine the lower range of K_d values for Pu and U.

1. Introduction.

The scope of work for this study had two overall objectives (Document #: CSM-02-97): 1) To provide a preliminary determination of the range in Pu phase speciation and soil distribution coefficient (K_d) values in 903 Pad area soils; 2) determine plutonium (Pu) inventories for ponds C-2, B-1 and B-5. This data, coupled with information about water flow during 'normal' rains

and storm events, will provide a basis for further evaluation of the rate of radionuclide transfer to surface waters and the link between surface water quality and soil action levels (see Honeyman and Santschi, *A conceptual model of Pu movement through RFETS soils*). The overall goal is to provide information as a foundation for assessing whether additional soil cleanup goals must be established, or further institutional controls that may be needed, for the protection of surface waters.

It has long been understood that the fate of an element in the environment depends on two factors: 1) the speciation of the element; 2) the processes that act to distribute those species. Establishing a link between surface water quality and soil action levels requires a thorough understanding of both elemental speciation and transport. The gross concentration (or activity) is important to a lesser extent.

The work summarized below, and covered under the Work Scope Document (Document #: CSM-02-97), falls into three categories:

1. Determination of the *phase speciation* of Pu in a) RFETS soils near the 903 Pad 'lip' area; b) South Interceptor Ditch (SID) sediments; c) Pond C-2.
2. An evaluation of the 'mass loading' of Pu to Pond C-2, B-1 and B-5.
3. Determination of K_d values for U(VI) interaction with Solar Pond soil materials.

2. Materials and Methods.

2.1 Selective leaching.

2.1.1 Materials evaluated.

Selective leaching analysis was performed on three types of RFETS media:

1) Soils near the 903 Pad 'lip' area;
2) South Interceptor Ditch (SID) sediments;
3) Pond C-2 sediments.

The three sites in the Woman Creek drainage represent successive stages in the transport of $^{239,240}Pu$ through the drainage. Soils near the 'lip' area are the source term, the SID contains sediments derived from the hillside to the north if the ditch and pond C-2 sediments are 'integrators' of Pu activity in the basin.

3

2.1.2 Sample locations and form.

Figure 1 shows the location of the samples at the time of extraction. All samples were provided to CSM and Texas A&M University, by Rocky Mountain Remediation Services. Soil samples 1 through 5 were taken from the 903 Pad area in a line extending toward SW 53. Sample 6 was taken from an 'alluvial fan', directly South of the 903 Pad. The soil samples were homogenates of 'box cores' (ca. 10 cm x 10 cm x 10 cm); as such, the soil analyses presented in Appendix 5 represent a homogenized surface soil (0 - 10 cm).

Three SID samples were analyzed and are designated SID 25, SID 27 and SID 39. The samples were homogenized, split into two 100 g sub-samples and frozen.

Two types of Pond C-2 samples were taken: 1) pond sediment homogenates for $^{239,240}Pu$ phase speciation and inventory estimates; 2) intact cores for sediment profiles of $^{239,240}Pu$. The Pond C-2 samples used for the phase speciation were C-2 #3 and C-2 #5.

2.1.3. ^{242}Pu yield tracers.

The ^{242}Pu used by Texas A&M as a yield tracer was NIST standard solution NIST SRM 4224F at 28.26 Bq g^{-1} with a relative expanded uncertainty (k = 2) of 0.74%. The dilution consisted of 1.46 g in 500 ml of 16% HNO_3 resulting in 4.98 ± 0.04 dpm ml^{-1}.

The ^{242}Pu used by CSM was NIST standard solution NIST SRM 4334F at 28.26 Bq g^{-1} with a relative expanded uncertainty (k = 2) of 0.74%.

2.1.3. Experimental protocol.

A number of selective extraction protocols have been proposed with many tailored to the specific substrates under study (see Table 1). This study followed the general scheme suggested by Yong *et al.* (1993) with a few modifications. The initial soil or sediment mass used in the extraction was 5 g. A detailed protocol is provided in Appendix 1.

Exchangeable cations:	8 ml KNO_3 at room temperature; agitation for 1 hour.
Carbonates:	8 ml of 1 M NaOAc adjusted to pH 5 with HOAc; agitation for 1 hour.
Sesquioxides:	20 ml of 0.04 $NH_2OH{\cdot}Cl$ in 25% (v/v) HOAc at 96°C for 6 h.
Organic matter:	Step 1: 3 ml of 0.02 M HNO_3 and 5 ml of 30% H_2O_2 adjusted to pH 2 with HNO_3 at 85 °C for 2 h; step 2: 3 ml of 30% H_2O_2 at

pH 2 and 85 °C for 3 hours with continuous agitation; Step 3: 5 ml of 3.2 M NH_4Oac in 20% (v/v) HNO_3 diluted to 20 ml at room T with agitation to 30 min.

Residual: Digestion with 5:1 mixture of HF and $HClO_4$; dissolve residue from digestion with 12 M HCl.

Additional variation of extraction parameters is described at the end of Appendix 5.

2.2 Pond sediment analysis.

2.2.1 Materials evaluated.

Two types of samples were provided to CSM and Texas A&M University by RMRS: 1) homogenized core samples; 2) intact cores.

2.2.2 Sample locations and forms.

Locations of the samples are given in Figure 1.

1. Intact cores. One intact core was taken from each pond for complete radionuclide profiles. Profile cores were frozen in an upright position to preserve the vertical structure near the sediment-water interface. Once frozen, the cores were extruded and cut in 1 cm intervals.

2. Core homogenates. Six core homogenates for radionuclide inventories and phase speciation were taken from each pond. Two of the six were sampling replicates. The bulk contents of the core (sediments plus porewater), including the layer near the interface, was emptied into a container and completely homogenized by stirring. During homogenization, core materials were in contact with the atmosphere. Two 100 gram aliquots from each inventory core were sent to CSM.

Appendix 2 contains the protocol for the extraction of $^{239,240}Pu$ from sediments. The plating procedure (the formation of a rare-earth precipitate) is given at the end of Appendix 3.

2.3 Distribution coefficients: 903 Pad 'lip' area soils.

2.3.1 Materials evaluated.

The soils used in the evaluation of soil/water partitioning coefficients, K_d values, are samples from the box-cores homogenates described in Section 1 and processed through a 2.36 mm sieve.

The solutions for the K_d experiments were supplied by RMRS: Seep water 491 and Well 1786 water.

2.3.2 Experimental protocol.

In these experiments, soil isolates (5 g of material < 2.36 mm) were suspended in 0.5 L of water (Seep: 491; Well 1786). The sieved fraction (i.e., the portion of the bulk soil smaller than 2.36 mm) represents between 10 and 20 % of the bulk soil by mass, depending on the sample location. The soil suspensions were shaken for five days, and in a few kinetics experiments for one, two and a half, and five days; the particles were then separated from solution and the solution analyzed for $^{239,240}Pu$. Each time point represents a separate batch reactor. A general definition of K_d is given below in Section 3. The experimental protocol followed for quantifying dissolved-phase $^{239,240}Pu$ activity is outlined in Appendix 3.

2.4. Distribution coefficients: solar pond cores and U(VI).

2.4.1 Materials evaluated.

Cores for the analysis were supplied by RMRS. Both cores (41193 and 54249) were significantly broken apart with the largest pieces being ca. 5 cm in length. RMRS also supplied the solutions for equilibration. Water #05193 is high in nitrate (sampled on 6.23.97 at 1430 h); water #1786 is low in nitrate (sampled on 6.23.97 at 1606 h).

2.4.2 Experimental protocol.

Appendix 4 gives the detailed experimental protocol followed for the K_d experiments. The procedure follows well-established protocols for evaluating trace element sorption through the use of a tracer (e.g., ^{233}U to trace ^{238}U).

3. Results and Discussion.

3.1. Phase speciation of Pu in RFETS environmental media.

3.1.1. General issues regarding phase speciation.

'Speciation' has different meanings, depending on the use and the authors of the studies. Speciation broadly means the 'form' of an element. However, 'form' may mean isotopic composition, physical form (e.g., gas, solid, a surface phase, etc.) or molecular composition (e.g., von Gunten and Benes, 1995). There are a number of techniques available for determining trace

element speciation; all have constraints associated with their use including volume and concentration limits, interference from non-target chemicals and the need for restrictive sample analysis environments (e.g., high vacuum). In the work described below, 'phase speciation' means the association of Pu with an operationally-defined phase or class of soil constituents.

The most frequently applied method for the analysis of trace element speciation of solid phases is selective leaching (sometimes also called sequential chemical extraction or phase speciation analysis). The methodology is to attack the soil or sediment with increasingly harsh chemical treatments, successively removing the target element from host soil or sediment components. Each of the chemical additions is targeted to destroy a particular class of phases, thereby releasing the 'bound' trace element. A number of different leaching schemes have been proposed over the years; the limitations of such techniques have been thoroughly discussed as well (e.g., von Gunten and Benes, 1995, and references therein). The main limitation of chemical leaching is the low selectivity of the applied chemicals for the target 'phases' (von Gunten and Benes, 1995), i.e.:

1. The extractants often do not quantitatively release the expected form(s), but also attack unwanted forms of the radionuclides;
2. The extractions may significantly change the abundances or properties of the unextracted components in the sample;
3. The extracted radionuclides can re-adsorb on the residue.

The interpretation of selective leaching results may be further complicated when the target metals may undergo redox changes upon application of host-phase extractants. For example, reductants added with the expectation that host mineral phases will reductively dissolve and thereby release the bound target trace metal may also, in some instances, reduce the target metal and alter the metal's association with soil mineral phases. In addition, the effectiveness of selective leaching analysis depends on:

1. Protocol characteristics including the length of time that extractants are in contact with the soil and the order of the various extraction steps;
2. The solid/solution ratio;
3. Soil structure.

It should also be noted that the results of selective leaching analysis provide only a 'snapshop' of the distribution of the target components and, unless supplemented by additional information, do not provide the basis for reaction processes and the future state of the soil or sediment system. However, in spite of the limitations of the approach, selective leaching can be a useful tool for comparing operationally-defined radionuclide speciation as a function of time and space within the same geochemical system.

Typically, the leach solutions are applied in series to a soil or sediment sample. Also, typically, five different 'phases' of the solid-phase associated trace elements are targeted: 1) an exchangeable fraction; 2) bound in carbonates; 3) associated with Fe and Mn (hydr)oxides or sulfides; 4) associated with organic matter; and 5) a residual fraction. The analyses presented below followed the sequence shown in Table 1. In this study, the reductant step preceded the peroxide addition in part to aid in evaluating the question of Pu release from the soil matrix under soil conditions which might foster the reductive dissolution of iron and manganese oxides (e.g., prolonged soil flooding).

Table 1 contains a comparison of some selected selective leaching schemes commonly applied to the analysis of soils and sediments. A more complete comparison of extraction procedures can be found in Yong *et al.* (1993). The work reported herein followed the general scheme of Yong *et al.* (1993), with a few modifications.

Table 2 summarized the results of selective leaching analysis. $^{239,240}Pu$ activities in the soil homogenates range from 372 pCi g^{-1} (Sample #2) to 2.6 pCi g^{-1} (Sample #5); total $^{239,240}Pu$ activity (e.g., pCi g^{-1}) in the homogenized surface soils generally decreases with distance from the 903 Pad.

The 'blank' soils were taken from an area near Coolbaugh Hall on the School of Mines campus in Golden, CO. Note that the purpose of the 'blank' is not to ascertain background $^{239,240}Pu$ fallout from nuclear weapons testing but for process control analysis of activity carryover between steps. Even though all analytical ware was washed in strong acid solutions between use, activity carryover still exists. The 'blanks' represent a maximum estimate of the carryover or 'blank' correction. Blank 1 is the correction for the high $^{239,240}Pu$ activity samples (soils 1 - 3) and Blank 2 is the corresponding correction for the low activity samples, soil samples 4 - 6, the SID samples and Pond C-2 homogenates. Thus, the activity carryover

correction for Soil #1 is $\frac{018}{246}$ x 100 or 0.072 %. For comparison, an average background soil $^{239,240}Pu$ activity for nuclear fallout is 0.05 pCi g^{-1}.

Table 1. Summary of selected extraction procedures.

Exchangeable	Bound in carbonates	Bound in Fe-Mn oxides	Bound in organic matter	Residual	Author
$MgCl_2$	NaOH/HOAc	$NH_2OH \cdot HCl$ in 25 % HOAc	H_2O_2/HNO_3 + NH_4OH	HF + $HClO_4$	1
$CaCl_2$	NaOAc; pH 5	CBD*	NaOCl; pH 9.2	pyrosulfate fusion	2
NaCl	HCl	EDTA + NH_4OH	acid digestion	--	3
$MgCl_2$	NaOH	HCl	acid digestion	--	4
KNO_3	NaOAc; pH 5	$NH_2OH \cdot HCl$	H_2O_2 (3 steps)	$HF/HClO_4$ + HCl	5

*Citrate, bicarbonate and dithionite.

1: Tessier *et al.* (1979); 2: Litaor and Imbrahim (1996); 3: Platter *et al.* (1992); 4: Cook *et al.* (1984); 5: Yong *et al.* (1993)

Duplicate analysis. 'Duplicate' analyses were performed on soils 1 - 4 and are designated 'D'. Duplicate analyses are as follows. Soil #1: straight duplicate; Soil #2: time of extraction; Soil #3: $MgCl_2$ versus KNO_3.

The dominant phase associations for Pu in all of the samples analyzed (soil, SID and pond) were the 'organic' fraction (range: 16% - 80%; average: 41.5%; standard deviation: 24.8) and the residual fraction (range: 18% - 77%; average: 54.5%; standard deviation: 23.9). The sequioxide fraction for the soil samples was typically less than 5% of the total. A comparison of $^{239,240}Pu$ activities progressing roughly down-gradient from the 'lip' area to Pond C-2 yields the following: 1) $^{239,240}Pu$ total activities decrease through the system; 2) the fraction of Pu in the organic 'phase' decreases, with organic fraction activities becoming similar to that of $^{239,240}Pu$ in the sesquioxide fraction. With few exceptions, the residual fraction is the dominant 'phase' association for Pu.

The selective extraction data for the 'sesquioxide' fraction suggests that the fraction of Pu in phases that would be subject to release upon the reductive dissolution of host phases, e.g., by prolonged saturation of soils, is small (from 1 to 5%). However, such a fraction if solubilized could significantly elevate the aqueous concentration of Pu and explain some of the observed surface water quality exceedances.

Pu association with organic matter may be significant because (e.g., Guillaumont and Adolf, 1992; Moulin *et al.*, 1992; Choppin, 1992; 1988; Nelson *et al.*, 1985): 1) organic matter can be soluble under a range of environmental conditions; 2) Pu/organic matter complexes are relatively strong; 3) organic matter may be involved in Pu redox chemistry.

Table 2. Summary of selective leaching analysis (pCi g^{-1}).

Soil	Exchangeable	Carbonate	Sesquioxide	Organic	Residual*	Total
1	0.13	0.35	5.4	94	146	246
1D	0.13	0.34	3.9	75	na	na
2	0.23	0.53	9.9	97	264	372
2D	0.18	0.41	5.4	87	na	na
3	0.029	0.063	1.5	63	14	78
3D	0.0049	0.060	1.3	15	16	32
4	0.010	0.034	1.1	6.4	17	24
4D	0.00050	0.03	0.98	4.3	6.2	12
5	0.0024	0.0092	0.12	1.6	0.85	2.6
6	0.0096	0.0093	1.6	4.2	20	26
SID #25	0.002	0.011	0.042	0.19	1.7	1.9
SID #27	0.0085	0.0080	0.070	0.27	1.8	2.2
SID # 39	0.0065	0.0056	0.0062	0.0059	0.1	0.12
C-2 #3	0.004	0.021	0.040	0.011	0.42	0.49
C-2 #5	0.004	0.016	0.12	0.15	2.1	2.4

*The operational nature of this approach is indicated by total organic carbon analysis of the residual fraction. Although the soil treatment designed to oxidize organic matter is relatively harsh, some organic matter survives and remains in the 'residual' fraction [1D: 0.32%; 2D: 0.57%; 3D 0.28%; 4D: 1.1 %; SID #39: 0.42%].

Litaor and Imbrahim (1996) conducted a selective leaching study of ^{239,240}Pu in 903 Pad 'lip' soils. They examined five soil cores from an area to the NW of SW 51 and evaluated the phase speciation of ^{239,240}Pu as a function of soil horizon. Their results indicate that a relatively small portion of ^{239,240}Pu exists in either the 'exchangeable' or 'carbonate' fractions (< 7 %) and that

the dominant phases for Pu are the organic, sesquioxide and residual fractions (56.2, 27.8 and 19.5%, respectively, on the average for the A horizon analyses). By comparison, this study yielded 41.5 and 55.5 % for the organic and residual fraction, respectively, for the six 'lip' area soils evaluated.

Marty *et al.* (1996) argue in their comment on the results of Litaor and Imbrahim (1996) that the interpretation of the results of selective leaching analysis of Pu in soils is complicated by the intricate nature of Pu redox chemistry. Marty *et al.* raise some important points regarding the strict interpretation of selective leaching analysis as to the 'location' of Pu species in soils. Their basic point is that attacks to the soil matrix by different oxidizing or reducing agents, and the subsequent release of Pu, may reflect alterations to Pu solubility through Pu redox reactions rather than destruction of host mineral phases, themselves.

Selective leaching strategies yield operational definitions of 'solubility' under imposed chemical conditions. For example, hydroxylamine hydrochloride has long been used to reductively dissolve iron and manganese oxides in soils and sediments as a part of compositional analysis for Fe and Mn. The application of an $NH_2OH{\cdot}HCl$ leach to phase speciation analysis should thus be limited to the interpretation that *under the conditions such that Fe and Mn would be reductively dissolved* a certain fraction of Pu will be released. Similarly, peroxide attack of soils, and the release Pu, are suggestive that Pu is associated with an organic phase. However, acidic peroxide solutions may reduce Pu to Pu(III), a relatively soluble oxidation state. Thus, all that is certain is that *under conditions sufficient for organic matter destruction* Pu is 'solubilized'. At this point, the significance of the 'organic' fraction for Pu remains unclear.

3.2. Pond ^{239,240}Pu activities.

Appendices 7, 8 and 9 contain the results of the analysis of Ponds B1, B5 and C-2 sediments. Appendix 7 contains the results of the homogenate analyses for Ponds B-1, B5 and C-2; Appendices 8 and 9 presents a summary of intact core analysis for cores B5-5 and C-2-1, respectively.

Pond B-5 sediments have specific activities ranging from 4.15×10^{-2} pCi g^{-1} (91.3 dpm kg^{-1}) to 0.58 pCi g^{-1} (1267 dpm kg^{-1}). Pond C-2 sediments exhibit a slightly larger range in specific ^{239,240}Pu activities: 8.27×10^{-1} pCi g^{-1} (182 dpm kg^{-1}) to 2.73 pCi g^{-1} (5984 dpm kg^{-1}). In

contrast, Pond B-1 has significantly greater $^{239,240}Pu$ specific activities than do either Ponds C-2 or B-5: 34.5 pCi g^{-1} (7.6×10^4 dpm kg^{-1}) to 111 pCi g^{-1} (2.44×10^5 dpm kg^{-1}).

Table 3 summarizes the homogenate results in terms of inventories (ΣpCi cm^{-2}) and includes duplicates. Duplicates are designated by A and B and consist of parallel analysis of identical sub-samples (in terms of mass) of the homogenates. Note that the reproducibility of duplicates is not typically high. Relatively low reproducibility was also noted by Litaor. Low reproducibility is suggestive of non-homogeneous dispersion of $^{239,240}Pu$ throughout environmental media. It is not clear at this time what the scale-length of homogeneity (e.g., 1, 5, 10 or more grams of media) is for $^{239,240}Pu$ in various environmental media or position within a media (i.e., sample location).

Pond inventories (mCi) were estimated by two different methods: 1) from the homogenates; 2) from core profiles. Both techniques should provide equivalent inventories. The inventories were calculated from the homogenates by assuming that the homogenate activity for each sediment location represents an average integrated activity, cm^{-2}, for the sample location. Inventories for each core homogenate (ΣpCi cm^{-2}) were calculated as follows:

$$\Sigma Pu\left(\Sigma pCi\ cm^{-2}\right) = Pu\left[dpm\ g^{-1}\ dry\ wt\right] \times (1-\theta) \times 2.5\left[g\ cm^{-3}\right] \times h[cm] \qquad (1)$$

where 2.5 is the dry density, θ the porosity and h the height of the homogenate section. The homogenate results were averaged (Table 3) and multiplied by the average pond area (Table 4) to yield the total pond inventory.

Intact cores were retrieved for Ponds B-5 and C-1. Appendices 8 and 9 contain the data summaries for the core analyses. Figure 2 presents the core $^{239,240}Pu$ profiles. The integrated core activities are presented in Table 5.

The last column in Appendix 7 contains the estimated 'integrated' $^{239,240}Pu$ activity per unit area of sediment/water interface. Pond B-1, again, has significantly greater integrated $^{239,240}Pu$ activities (1870 pCi cm^{-2}) than does either Pond C-2 (12 pCi cm^{-2}) or B-5 (3.3 pCi cm^{-2}).

Analyses of intact cores for $^{239,240}Pu$ are given in Appendices 6 and 7. Note that Pu specific activities are highly variable in both sets of cores, with Pond C-2 exhibiting a higher average specific $^{239,240}Pu$ activities throughout the core. Note, also, that the Pu activities at the base of the Pond B-5 core are significantly less than are activities in the upper half of the same core.

12

The $^{239,240}Pu$ activity in each core slice is given by

$$\Sigma Pu(pCi\ cm^{-2}) = Pu[dpm\ g^{-1}] \times \frac{1}{2.2}\left[\frac{pCi}{dpm}\right] \times (1-\theta) \times 2.5[g\ cm^{-3}] \times 1\ cm \qquad (2)$$

The integrated core activity is found by summing all Pu activities including interpolated values. The integrated core activities are: 14.4 pCi cm^{-2} and 30.7 pCi cm^{-2} for cores B5-5 and C2-1, respectively.

Average pond inventories range from 0.5 to 42 mCi of $^{239,240}Pu$ for Ponds B1 and B5, respectively, when calculated from core homogenates (Table 3); inventories are 2 to 5 times higher when calculated from the more reliable vertical core profiles (Table 5). The reason for this difference could be in the shallow penetration of the cores taken for the homogenates.

Average yearly fluxes to Ponds B1, B5 and C-2 are listed in Table 6 and range from 10^{-2} to 10^{0} mCi y^{-1}, depending on the pond and method of integration. These values could be used to calculate soil-Pu mechanical erosion rates in the drainage basins for these ponds. The yearly fluxes were calculated from the following pond age information: 1) B-1 was constructed in 1972 (Δt = 25 y); 2) B-5 was constructed in 1978 and had a major modification of the outlet works in 1984 (Δt = 13 y); 3) C-2 was constructed in 1978 (Δt = 19 y).

^{137}Cs activity. ^{137}Cs, derived from bomb fallout during atmospheric weapons testing, has been used to date sediment deposition in a number of systems (e.g., Santschi and Honeyman, 1989, and references therein) and to provide information on uniformity of sediment deposition. ^{137}Cs activities in pond core slices were too low to provide useful relative counting errors.

Table 3. Summary of homogenate results.

Pond	Average Integrated Activity (ΣpCi cm^{-2})	Average $^{239,240}Pu$ in Pond (mCi)
B-1	1870	42.4
B-5	3.8	0.45
C-2	12.1 (21.7)[a]	2.42

[a]Corrected to penetration depth of profile core (core analysis only for 10 cm instead of 18 cm in C-2).

13

Table 4. Summary of pond areas (acres*).

Pond	Minimum	Maximum	Average
B-1	0.35	0.68	0.56
B-5	1.44	4.84	2.96
C-2	2.59	6.47	4.95

*There are 4.05×10^7 cm^2 $acre^{-1}$.

Table 5. Summary of core inventories.

Pond	Integrated Activity (ΣpCi cm^{-2})	Average $^{239,240}Pu$ in Pond (mCi)
B-5	14.4	1.72
C-2	30.7	6.15

Table 6. Estimation of $^{239,240}Pu$ yearly activity flux to ponds (mCi y^{-1})[a].

Pond	From Homogenates	From Cores	Pond Age (y)
B-1	1.70	na[b]	25
B-5	0.024	0.13	13
C-2	0.13 (0.22)[c]	0.32	19

[a]The fluxes were calculated using the pond ages given in the text.
[b]The integrity of the Pond B-1 core was lost during shipment.
[c]Corrected to penetration depth of profile core (core analysis only for 10 cm instead of 18 cm in C-2).

3.3. K_d values for $^{239,240}Pu$ in lip soils and U(VI) with solar pond cores.

3.3.1. The significance of K_d values.

Mathematical models for simulating the transport of chemical constituents through soils by water typically treat the transport as occurring primarily by the dissolved form. However, it is rare to find an element that resides solely in the water (or dissolved) phase. Typically, a dissolved element will interact to some extent with the particle (and usually immobile) phase.

14

The partitioning of an element between the dissolved and particle phases is often described through the use of a distribution coefficient, K_d. For a radioactive element, the K_d is simply the ratio of the activity concentration of an element in the particle phase, e.g., pCi kg^{-1}, to the corresponding activity concentration in the 'dissolved' phase, pCi L^{-1}. As a result, the dimensions of K_d are usually L kg^{-1}. As a consequence of the empirical definition of K_d, no relationship to thermodynamically defined partition coefficients is implied as the empirical definition includes not only surface sorption control but also other mechanisms such as solubility control by oxides and colloids that partition into the solution phase.

A distribution coefficient reflects the net of all chemical and physical processes that result in the distribution of an element between the particle and solution phases. A low K_d element partitions strongly to the water; a high K_d element partitions more strongly to the particle phase. Examples of low K_d species are Na^+ (the sodium ion) and HCO_3^- (bicarbonate). Because transport through soils primarily occurs as the result of water movement (or transport 'through' the dissolved phase), a high K_d element will not be very mobile as a consequence of dissolved-phase transport. The variability in K_d values for a particular element reflects the range in environmental conditions at the site where the K_d values will be applied. As such, K_d values are 'conditional' on the characteristics of the system: K_d values simply reflect the empirical distribution of a radionuclide between a solid phase and a water phase that is in contact with the solid. A number of discussions on the relationship between system chemistry and K_d values are available (e.g., Davis and Kent, 1990, and references therein).

Most computer models for simulating the fate of radionuclides in the environment use K_d values as part of the 'mobility' calculations in the model. While empirically-defined K_d values do not represent fundamental chemical characteristics of a system (i.e., they are not 'state variables') they are quite useful, to the extent that their variability can be quantified, in evaluating the range of mobilities that can be expected in a target environment for the element of interest.

As an example of the conditional nature of K_ds, it is well established that the partitioning of Pu (and many other trace metals) between particles and the water phase depends on the concentration of organic matter that is present in the water phase (e.g., Berry *et al.*, 1991; Choppin, 1988; Lenhart, 1997). K_d values for Pu partitioning to soil particles may decrease in

15

value as the concentration of dissolved organic in the soil water increases. Organic matter 'shifts' the partitioning of Pu from the particle to the water phase because, under certain circumstances, Pu more strongly associates with dissolved organic matter than with particles. Because K_d values are not typically based upon a knowledge of speciation (e.g., the formation of chemical compounds between Pu and dissolved organic matter), but are specific to a certain set of environmental conditions, it is usually not possible to quantify how a K_d value will change in response to variations in system conditions. As such, a soil system in which the concentration of dissolved organic matter changes with storm event intensity will have 'system' K_d values for radionuclide partitioning to particles that also change with storm event intensity.

Understanding the range in the expected particle/water partitioning of an element is important because the extent of partitioning regulates the mobility of an element by transport through the aqueous phase. More explicitly, the K_d is related to the transport velocity of an element, relative to the velocity of the water, itself. For example, a compound with a K_d of $\approx$ 0 L kg^{-1} will move at the same velocity as the water phase; a compound with a K_d of 1000 L kg^{-1} will move approximately at $1/1000^{th}$ the velocity of the carrier phase (i.e., the water).

One assumption that is often made when using K_d values in mobility calculations is that a chemical equilibrium exists in the partitioning of an element between particle and dissolved phases. Depending on the form of the dissolved Pu this assumption may not be valid. For example: 1) partitioning (i.e., the movement of Pu from the dissolved phase to the particle phase or the reverse) may be slow; 2) the 'dissolved' Pu may not be truly dissolved but may be in the form of micro-particles (i.e., colloids[1]), which are often operationally defined as part of the dissolved phase (colloid-facilitated transport of contaminants require adaptations to transport codes typically used for dissolved-phase transport, e.g., Reible *et al.*, 1991); or 3) a significant portion of the particle-phase Pu activity may not be available for exchange into the water phase (e.g., the 'residual' fraction).

3.3.2. *K_d values for $^{239,240}Pu$ in lip soils.*

The K_d values were determined by a mass balance on $^{239,240}Pu$

[1] Colloidal materials are typically defined as substances that will pass a 0.45 μm filter but which have a molecular weight of 1000 daltons (atomic mass units) or more (about 10^{-9} m and larger).

$$K_d = \frac{Pu_{particulate}\left(dpm\ kg^{-1}\right)}{Pu_{solution}\left(dpm\ L^{-1}\right)}\left[L\ kg^{-1}\right] \quad (3)$$

Table 7 is a list of K_d values for $^{239,240}Pu$ in equilibrium with 903 Pad 'lip' area soils. Appendix 10 contains the complete analytical data. Note that the distribution coefficients are based on the activity of $^{239,240}Pu$ that was transferred from the soil particles to solution over the 5 day experiment. Solutions were in equilibrium with atmospheric CO_2. The K_d values most likely represent a lower limit because: 1) the suspensions were stirred to increase mass transfer; and 2) of the high solution to solid ratio.

Table 7. List of $^{239,240}Pu$ K_d values obtained for 903 Pad 'lip' area soils.

Sample	Water #	K_d (L kg^{-1})	Error (1 σ)	Final pH
971879 1	491	0.98×10^4	0.04×10^4	8.39
971879 2	491	1.16×10^4	0.05×10^4	8.30
971879 3	491	3.59×10^4	0.33×10^4	8.40
971879 4	491	8.7×10^4	1.3×10^4	8.39
971879 5	491	1.35×10^4	0.45×10^5	8.21
971879 6	1786	1.16×10^5	0.09×10^5	7.94

The relationship between soil-phase activity and the dissolved-phase $^{239,240}Pu$ in 'instantaneous' equilibrium with the solid phase is given by

$$f_{dissolved} = 1 - \frac{K_d C_p}{1 + K_d C_p} \quad (4)$$

where $f_{dissolved}$ is the fraction of $^{239,240}Pu$ (particulate + dissolved) that is in the solution phase and C_p is the particle concentration (kg L^{-1}). If, for example, 5 grams of the soil isolate used in the K_d experiments (i.e., the < 64 μm fraction) are in contact with 1 L of groundwater ($^{239,240}Pu_T$ = 1.64 x 10^2 pCi g^{-1}; K_d = 1 x 10^4 L kg^{-1}), the resulting dissolved $^{239,240}Pu$ would be 3.32 pCi L^{-1}. Assuming that Pu is only in the fine fraction (< 2.33 mm; an assumption that must be evaluated), measured K_d values can be scaled to the total soil mass by only considering fine-grained material as the carrier.

17

3.3.3. Desorption kinetics.

Figure 3 shows K_d values for Pu release from soil isolate 97L1879 #2 as a function of time and water type. K_d values are dependent on both the composition of the water in contact with soil particles and time. It is clear that, for the system containing water from well 1586, equilibrium partitioning has not been reached.

3.3.4. Solar pond soils and U(VI).

Appendix 11 presents the results of K_d evaluations for the interaction of U(VI) with solar pond soil isolates; Appendix 4 contains the experimental protocol. The background solutions for the analysis were a 'high' nitrate (water # 05193) and a 'low' nitrate (water # 1786). The core isolates used in the sorption experiments constitute the less than 64 μm particle-size fraction; consequently, the K_d values derived in this study represent an upper limit relative to what would be observed for 'bulk' core K_d values. The reported K_d values represent 64 hours of contact between the soil particles and solutions. The pH values were recorded after the 64 hours of equilibration. Appendix 12 contains a reproduction of the borehole logs.

The sorption data (Table 8) exhibit three broad characteristics: 1) K_d values that range over nearly an order of magnitude (from 31.2 to 148.7 L kg^{-1}); 2) K_ds that decrease in value from core top to bottom; and 3) relatively little influence of solution composition on the magnitude of U(VI) sorption. The magnitude of the K_d values is consistent with the relatively greater solubility of U(VI) with respect to sorptive processes, compared to other actinides. The greater solubility is the consequence of generally weaker interactions between U(VI) and mineral surfaces and the formation of non-sorbing or weakly-sorbing solution-phase U(VI) complexes (such as uranyl carbonato complexes) (e.g., Choppin, 1988, 1992; Lenhart *et al.*, 1997). The decrease in K_d values with depth reflects changes in the composition of the sorption substrate and is possibly due to a decrease in the ratio of silica (sand) to clay down-core.

3.3.5. Sorption kinetics.

Sorption kinetics data for $^{233}U/^{238}U$ interaction with solar pond core material yielded no statistically significant difference in K_d values over a six day experiment period. Core materials considered were: 1) top, core 54294 and 2) top, core 41193. The water used for the suspension of the core material was the low nitrate water, #1786.

18

Table 8. Summary of K_d values for U(VI) sorption onto Solar Pond cores.

Core 54294:

Depth	Nitrate	K_d (L kg^{-1})	Average K_d
t	h	153.3	
t	h	145.9	
t	h	146.8	148.7 ± 4.0
t	l	173.1	
t	l	172.1	
t	l	167.9	171.0 ± 2.8
m	h	55.6	
m	h	57.1	
m	h	54.5	55.8 ± 1.3
m	l	58.2	
m	l	60.1	
m	l	58.6	59.0 ± 1.0
b	h	31.9	
b	h	29.4	
b	h	32.4	31.2 ± 1.6
b	l	45.3	
b	l	44.4	
b	l	61.8	50.5 ± 9.8

Core 41193:

Depth	Nitrate	K_d (L kg^{-1})	Average K_d
t	h	111.7	
t	h	103.2	
t	h	109.7	108.2 ± 4.5
t	l	118.4	
t	l	123.3	
t	l	128.3	123.4 ± 4.9
m	h	50.4	
m	h	43.8	
m	h	51.7	48.6 ± 4.2
m	l	60.3	
m	l	54.2	
m	l	51.1	55.2 ± 4.6
b	h	55.7	
b	h	55.5	
b	h	62.4	57.7 ± 4.0
b	l	64.0	
b	l	67.2	
b	l	50.6	60.6 ± 8.8

Nomenclature.

Core 54294. t (top): 7.8 - 9.0 ft; m (middle): 9 - 10 ft; b (bottom): 12.5 - 13 ft.
Core 41193. t (top): 4.3 - 5.1 ft; m (middle): ca. 6.9 ft; b (bottom): ca. 7.6 ft.
High nitrate: 05193
Low nitrate: 1786

19

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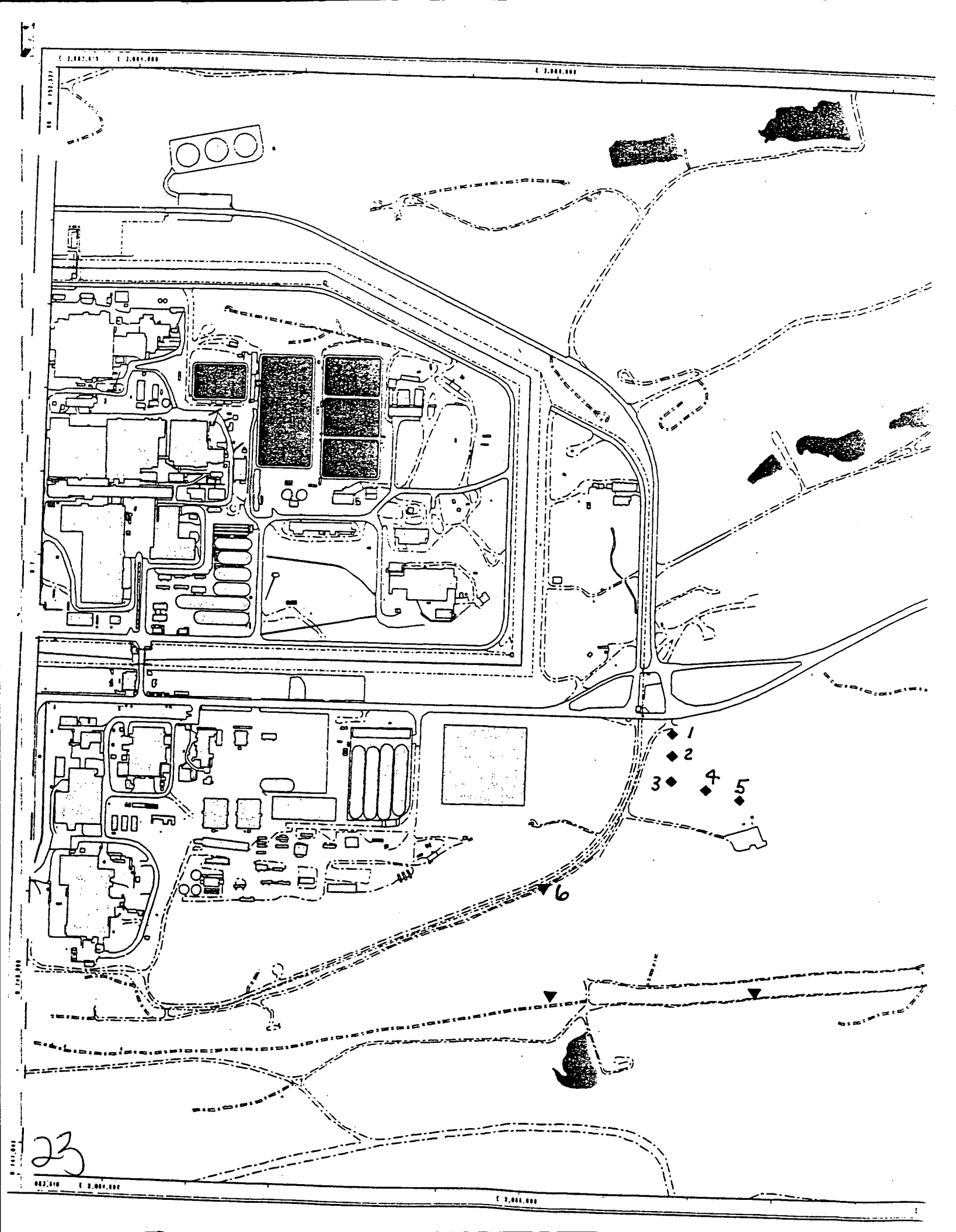
21

5. List of Figures.

Figure 1. Location of samples taken for analysis at Rocky flats Environmental Technology Site.

Figure 2. $^{239,240}Pu$ activity (pCi g^{-1}) *versus* depth (cm) in pond cores. a) Core B5-05; b) Core C2-01. The line drawn through the data is presented as an aid in visualizing the structure of the activity *v.* depth profile and is not a mathematical fit to the data.

Figure 3. K_d (L kg^{-1}) as a function of time (days) and water type for $^{239,240}Pu$ desorption from soil isolate 97L1879# xxxx.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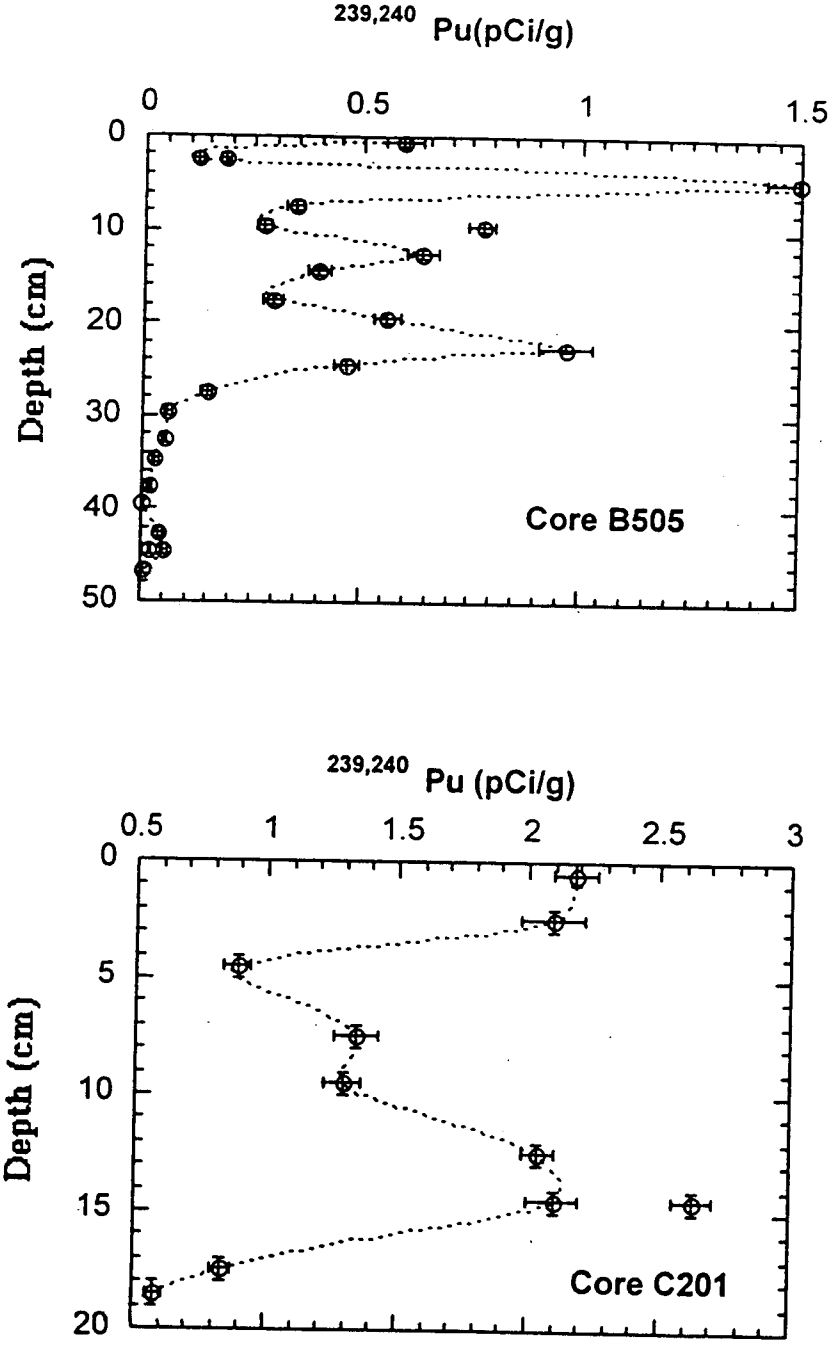

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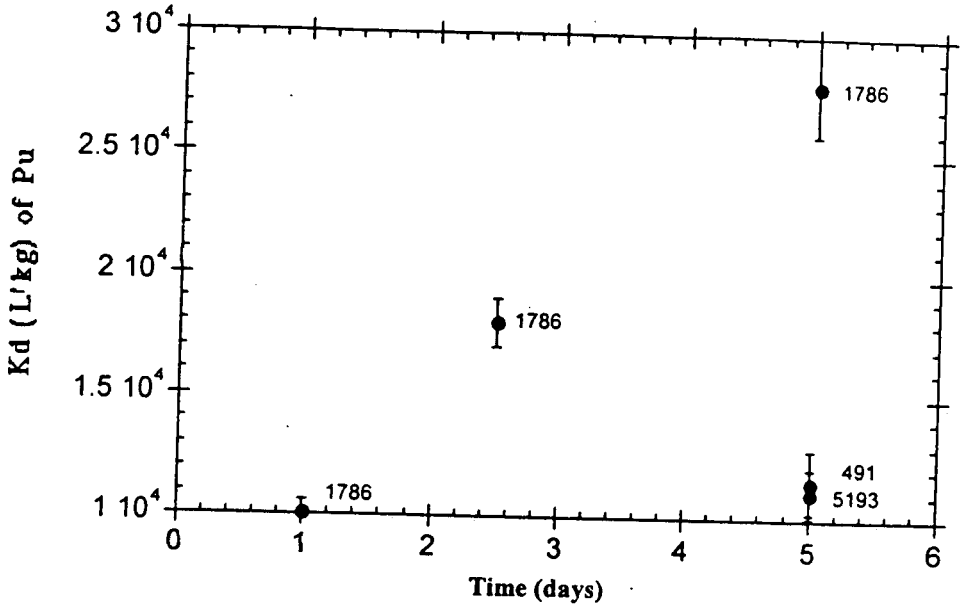

Figure 3. K_d (L kg^{-1}) as a function of time (days) and water type for $^{239,240}Pu$ desorption from soil isolate 97L1879# 2.

6. Appendices.

1. Analysis procedure for Rocky Flats soils/sediments in concert with selective extraction for phase speciation.
2. Protocol for Pu extraction from sediments.
3. Protocol for Pu extraction from water.
4. Protocol for the determination of partitioning coefficients for U(VI) interaction with solar pond soil core.
5. Results of sequential leaching.
6. Carbon analysis.
7. Summary of pond core homogenates.
8. Core B5-5 analysis.
9. Core C2-1 analysis.
10. $^{239,240}Pu$ K_d values for 903 Pad 'Lip' area soils.
11. U(VI)/solar pond core K_d values.
12. Borehole logs for solar pond cores 41193 and 54294.

APPENDIX 1: ANALYSIS PROCEDURE FOR ROCKY FLATS SOILS/SEDIMENTS IN CONCERT WITH SELECTIVE EXTRACTION FOR PHASE SPECIATION (VER. 1.0)

Transfer a quantity of sediment that has been semi-dried and sieved through a 2-3 mm mesh screen, to a dish for drying.

Place the dish containing the sediment into a low heat ventilated oven (<80 degrees C) until the weight is stable after cooling to room temperature.

Weigh out 3.0 +/- 0.1 grams of sediment into well sealing, 50 ml, conical bottom centrifuge tube.

Prepare a known blank sediment with each batch of sediments to be extracted.

Duplicate one of the samples in the batch to run exactly as its duplicate.

Duplicate one of the samples to run with excessive extraction times for comparison to its duplicate.

Duplicate one of the samples to compare the exchangeable extractants - potassium nitrate vs. magnesium chloride.

Determine the quantity of extractant used to the nearest tenth of a milliliter.

Follow the extraction scheme as detailed in Yong *et al.* (1993), with the following additions:

> After the prescribed extraction time, separate the extractant from the sediment by centrifugation and decantation followed by filtration of the decanted liquid through a 0.45 micron membrane filter.
>
> Determine the quantity of extractant left in the sediment by re-measuring the extractant recovered.
>
> Rinse any residues on the filter back into the tube containing the majority of the sediment with approximately three times the quantity of residual extractant using D.I. water as the rinse.
>
> Vortex the sample for several seconds, recentrifuge and filter the decanted supernate through a 0.45 micron membrane filter and combine with the original extractant.
> (Proceed with the next extractant, repeating this sequence.)

Spike the extractant with a known amount of ^{242}Pu. Use approximately one-tenth the expected quantity of $^{239,240}Pu$ in the entire extracted sediment. (e.g., a sample is 100 pCi/g $^{239,240}Pu$. A 3 gram extraction aliquot is taken. The Pu-242 tracer used should be 30 pCi for each extraction.)
For extractants 1-4:

Add 10 ml conc. nitric acid slowly. Allow to stand for several minutes at room temperature, then place on a warm hot plate for several minutes. Add a few drops of hydrogen peroxide to assist with the destruction of organic material, extractant residue and to assist in the exchange of tracer with the analyte.

Take the solution to dryness on a moderately hot hotplate. If it appears that the residue will not redissolve due to insoluble material repeat the above treatment with nitric acid and hydrogen peroxide.

For extractant 5:

Rather than adding the 12 M hydrochloric acid and diluting as in the "Yong paper", take the acid mixture to the point of perchloric acid fumes (dense white clouds) for a few minutes, being careful not to allow the residue go to dryness. Add a small amount of perchloric acid if necessary to prevent going to dryness.

Allow the sample to cool then proceed with the following.

All samples:

Add 1-3 ml of conc. nitric acid and warm the solution on a hot plate.

Dilute the solution to 10 ml using 1M nitric acid.

Add 1 ml of 20 mg/ml Fe(III) carrier.

Add a spatula tip full of sodium nitrite crystals to the solution and mix well.

Allow the sample to stand for at least ten minutes.

Add conc. ammonium hydroxide in excess to precipitate the Fe carrier as iron hydroxide.

Centrifuge the sample for 5-10 minutes and discard the supernate.

Estimate the volume of the ferric hydroxide precipitate, add 4 times the volume of conc. hydrochloric acid and mix by vortexing. Add 9N hydrochloric acid to bring the soution to a final volume of 10-15 ml.

Fill a disposal plastic column (approximately 5ml I.D., 10 ml capacity) with 2 ml of AG 1X8 anion exchange resin. Cover the top of the resin with a small plug of glass wool.

Condition the resin with 10 ml of 9N hydrochloric acid. Discard the effluent.

Filter the solution onto the column if necessary.
Load the sample solution onto the column. Discard column effluent in appropriate waste

28

stream.

Rinse the column with 1,2,5,10 ml successive rinses of 9N hydrochloric acid allowing each rinse to pass completely through before adding the next.

Rinse the column with 3 successive 5 ml portions of 0.5 N nitric acid. Collect these portions in a plastic test tube.

Add approximately 0.5 ml 30% hydrogen peroxide and swirl.

Add 100 µg lanthanum carrier. Mix well. Add 5ml 25% HF. Mix well. Allow the sample to stand for 15-20 minutes.

Place a 25 mm 0.1 µm filter membrane in a filter funnel assembly and wet the membrane with a small amount of methanol or ethanol. Vacuum filter the solution then rinse with 10-15 ml of slightly basic water.

Remove the filter, dry at low heat, and mount on the planchet with double coated tap for APHA (alpha pulse height analysis).

Citations.

Yong, R.N., R. Galvez-Cloutier and Y. Phadungchewit (1993). Selective sequential extraction analysis of heavy-metal retention in soil. *Can. Geotech. J.*, **30**, 834-847.

APPENDIX 2: PROTOCOL FOR PU EXTRACTION FROM SEDIMENTS.

Preparing Sample.

1. Weigh 1.0 g sediment
2. Add spike (^{242}Pu)
3. Add 10ml conc. HNO_3
4. Add 10ml conc. HF
5. Let solution sit overnight
6. Microwave digestion

Procedure.

1. Transfer solution to a teflon beaker
2. Add 5ml perchloric acid; evaporate
3. Add 10ml con HNO_3, 100ml DH_2O +2g H_3BO_3 ; heat ~15min
4. Transfer to a 250ml centrifuge bottle; add 2ml $FeCl_3$
5. Add 1ml $NaNO_2$, vortex, then warm on hot plate for 15-30min
7. Add 40-50ml conc NH_4OH to form a precipitate (basic)
8. Centrifuge 15min- discard supernatant
9. Estimate volume of solid, add 3X amount with conc. HCl; vortex.
10. Add 75ml 9M HCl, then add 2ml $NaNO_2$, mix well, then set aside for 15min

IMPORTANT: If foamy gel present, perform PEG treatment (step 11)

11. Add 2-5ml 0.25M polyethylene glycol, vortex and set aside 30min, then centrifuge 5min

Column Preparation:

1. Fill column to 7cm w/resin
2. Place glass beads on top of resin (~2cm)
3. Place plastic funnel on columns, fold a Whatman 41 filter paper inside funnel, wet w/ 9M HCl
4. Condition resin w/50ml 9 M HCl, discard

Columns:

1. Load sample through filter in funnel
2. Rinse column 4x w/20 ml 9M HCl; allow each rinse to pass completely though before the next rinse is poured, discard effluent
3. Ellute column w/20 ml 9M HCl + 1.5ml HI; collect elluent
4. Add 1ml conc. HNO_3; evaporate to dryness

29

APPENDIX 3: PROTOCOL FOR PU EXTRACTION FROM H_2O.

Sample preparation.

1. Weigh 5.0 g sediment
2. Add 500ml H_2O (well/pond)
3. Stir for 5 days

Procedure.

1. Transfer solution and centrifuge ~15min
2. Collect supernatant, filter through 0.4 micron nuclepore filter paper
3. Collect filtrate, add 1ml ^{242}Pu tracer, and 10 ml conc. HNO_3
4. Set on hotplate to evaporate
5. Estimate volume of solid, add 3X amount with conc. HCl, vortex with conc. HCL, vortex
6. Add 75ml 9M HCl, then add 2ml $NaNO_2$, mix well, then set aside for 15min

IMPORTANT: If foamy gel present, perform PEG treatment (step 11).

7. Add 2-5ml 0.25M polyethylene glycol, vortex and set aside 30min, then centrifuge 5min

Column Preparation:

1. Fill column to 7cm w/resin
2. Place glass beads on top of resin (~2cm)
3. Place plastic funnel on columns, fold a Whatman 41 filter paper inside funnel, wet w/ 9M HCl
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Columns:

1. Load sample through filter in funnel
2. Rinse column 4x w/20ml 9M HCl; allow each rinse to pass completely though before the next rinse is poured, discard effluent
3. Elute column w/20ml 9M HCl + 1.5ml HI; collect eluent
4. Add 1ml conc. HNO_3; evaporate to dryness

30

Plating:

1. Add 1ml conc. HCl, mix well, then add 14ml D H_2O, mix well
2. Add 1ml lanthanum carrier + 0.5ml H_2O_2; mix, then add 5ml 3M HF; mix
3. Let sample stand 15-20min
4. Set up filtration apparatus
5. Apply vacuum; rinse filter w/methanol, then rinse w/ D H_2O
6. Transfer sample through filter, then rinse beaker w/5ml D H_2O, add to filter
8. After sample has passed through filter, rinse filter w/10-15ml D H_2O
9. Collect filter paper, mark paper w/sharpie
10. Mount filter paper on 1.25" planchet

31

APPENDIX 4: PROTOCOL FOR THE DETERMINATION OF PATITIONING COEFFICIENTS FOR U(VI) INTERACTION WITH SOLAR POND SOIL CORE ISOLATES.

1. 10.00 g of the 200 mesh core isolates was weighed into a vial.
2. The soil was quantitatively transferred from the vial to polycarbonate 'Oak Ridge' type centrifuge tubes.
3. 25 ml of 'high' or 'low' nitrate water was added to each tube.
4. 0.46 ml of ^{238}U stock solution and 0.500 ml of ^{233}U stock solution was added to each tube. U_T = 1 mM.
5. Each tube was mixed by tumbling for 64 h.
6. At 10 h intervals, tubes were opened to equilibrate with atmospheric CO_2.
7. At the end of the equilibration period, samples were centrifuged at 9000 3000 relative centrifugal force (rcf) for 30 min.
8. The pH of the supernatent liquid was measured.
9. 2 ml of the supernatent liquid was pipeted into vials for liquid scintillation analysis.

APPENDIX 5. RESULTS OF SEQUENTIAL LEACHING.

Sample	Fraction	Mass (g)	Pu-242 (dpm)	Tracer Counts	Pu-239/240 Counts	Count Time (m)	Sample Activity (dpm/g)	Counting Uncertainty (1σ)(dpm/g)	Sample Activity (pCi/g)	Counting Uncertainty (1σ)(pCi/g)	Σ(Activity) (pCi/g)	Percent of Total (%)
97L1879-001	exchangeable	3.05	13.63	3472	232	1000	0.30	0.02	0.13	0.01		0.05
97L1879-001	carbonate	3.05	9.08	1429	370	1000	0.77	0.04	0.35	0.02		0.14
97L1879-001	sesquioxide	3.05	8.82	359	1490	150	12	1	5.4	0.3		2.2
97L1879-001	organic	3.05	8.86	284	20295	150	208	12	94	6		38
97L1879-001	residual	3.05	16.7	1159	68818	500	325	10	146	4	246	60
97L1879-001D	exchangeable	3.07	13.63	3745	246	1000	0.29	0.02	0.13	0.01		
97L1879-001D	carbonate	3.07	9.08	2628	675	1000	0.76	0.03	0.34	0.01		
97L1879-001D	sesquioxide	3.07	8.86	348	1031	150	8.6	0.53	3.9	0.2		
97L1879-001D	organic	3.07	8.81	288	16807	154	167	10	75	4		
97L1879-001D	residual	3.07			lost tracer							
97L1879-002	exchangeable	3.05	18.17	3964	343	1000	0.52	0.03	0.23	0.01		0.06
97L1879-002	carbonate	3.05	9.08	2618	1043	1000	1.19	0.04	0.53	0.02		0.14
97L1879-002	sesquioxide	3.05	8.83	352	2677	150	22	1	9.9	0.6		2.7
97L1879-002	organic	3.05	8.76	85	6403	30	216	24	97	11		26
97L1879-002	residual	3.05	16.7	1123	120243	500	586	18	264	8	372	71
97L1879-002D	exchangeable	3.07	18.17	3577	240	1000	0.40	0.03	0.18	0.01		
97L1879-002D	carbonate	3.07	9.08	2756	838	1000	0.90	0.04	0.41	0.02		
97L1879-002D	sesquioxide	3.07	8.82	387	1616	150	12	1	5.4	0.3		
97L1879-002D	organic	3.07	8.86	146	9741	55	193	16	87	7		
97L1879-002D	residual	3.07			lost tracer							
97L1879-003	exchangeable	2.99	13.63	2867	40	1000	0.064	0.010	0.029	0.005		0.04
97L1879-003	carbonate	2.99	9.08	2439	113	1000	0.14	0.01	0.063	0.006		0.08
97L1879-003	sesquioxide	2.99	8.81	645	724	270	3.3	0.2	1.5	0.1		1.9
97L1879-003	organic	2.99	8.79	429	20289	150	139	7	63	3		80
97L1879-003	residual	2.99	16.7	27	147	500	30	6	14	3	78	18

Sample	Fraction	Mass (g)	Pu-242 (dpm)	Tracer Counts	Pu-239/240 Counts	Count Time (m)	Sample Activity (dpm/g)	Counting Uncertainty (1σ)(dpm/g)	Sample Activity (pCi/g)	Counting Uncertainty (1σ)(pCi/g)	Σ(Activity) (pCi/g)	Percent of Total (%)
97L1879-003D	exchangeable	2.99	13.63	3772	9	1000	0.011	0.004	0.0049	0.0016		0.02
97L1879-003D	carbonate	2.99	9.08	2567	112	1000	0.13	0.01	0.060	0.006		0.18
97L1879-003D	sesquioxide	2.99	8.82	641	649	270	3.0	0.2	1.3	0.1		4.14
97L1879-003D	organic	2.99	8.81	350	3995	154	34	2	15	0.8		46.67
97L1879-003D	residual	2.99	33.53	2588	8146	500	35	1	16	0.4	32	48.98
97L1879-004	exchangeable	2.97	9.08	2047	15	1000	0.022	0.006	0.010	0.003		0.04
97L1879-004	carbonate	2.97	9.08	2502	62	1000	0.076	0.010	0.034	0.004		0.14
97L1879-004	sesquioxide	2.97	8.8	648	537	270	2.5	0.1	1.1	0.1		4.5
97L1879-004	organic	2.97	8.85	2385	11357	1000	14	0.3	6.4	0.1		26
97L1879-004	residual	2.97	16.7	22	147	500	38	9	17	4	24	69
97L1879-004D	exchangeable	3.04	9.08	2687	1	1000	0.0011	0.0011	0.00050	0.00050		0.00
97L1879-004D	carbonate	3.04	9.08	2797	72	1000	0.08	0.01	0.03	0.00		0.3
97L1879-004D	sesquioxide	3.04	8.75	617	468	270	2.2	0.1	0.98	0.06		8.5
97L1879-004D	organic	3.04	8.84	2566	8468	1000	9.6	0.2	4.3	0.1		37.36
97L1879-004D	residual	3.04	33.53	3405	4268	500	13.8	0.3	6.2	0.1	12	53.83
97L1879-005	exchangeable	3.04	4.54	1135	4	1000	0.0053	0.0026	0.0024	0.0012		0.09
97L1879-005	carbonate	3.04	9.08	2621	18	1000	0.021	0.005	0.0092	0.0022		0.35
97L1879-005	sesquioxide	3.04	8.76	1805	169	800	0.27	0.02	0.12	0.01		4.6
97L1879-005	organic	3.04	8.84	2379	2985	1000	3.6	0.1	1.6	0.05		63
97L1879-005	residual	3.04	16.7	1866	642	500	1.9	0.1	0.85	0.04	2.6	32
97L1879-006	exchangeable	3.03	4.54	1125	16	1000	0.021	0.005	0.0096	0.0024		0.04
97L1879-006	carbonate	3.03	9.08	2612	18	1000	0.021	0.005	0.0093	0.0022		0.04
97L1879-006	sesquioxide	3.03	8.78	2040	2536	800	3.6	0.1	1.6	0.05		6.2
97L1879-006	organic	3.03	8.83	1548	5001	640	9.4	0.3	4.2	0.1		16
97L1879-006	residual	3.03	16.7	2082	16889	500	45	1	20	0.5	26	77

Sample	Fraction	Mass (g)	Pu-242 (dpm)	Tracer Counts	Pu-239/240 Counts	Count Time (m)	Sample Activity (dpm/g)	Counting Uncertainty (1σ)(dpm/g)	Sample Activity (pCi/g)	Counting Uncertainty (1σ)(pCi/g)	Σ(Activity) (pCi/g)	Percent of Total (%)
Blank 1 Soil	exchangeable	3.06	4.54	1028	1	1000	0.0014	0.0014	0.00065	0.00065		0.36
Blank 1 Soil	carbonate	3.06	9.08	2033	1	1000	0.0015	0.0015	0.00066	0.00066		0.36
Blank 1 Soil	sesquioxide	3.06	8.74	1659	4	800	0.0069	0.0034	0.0031	0.0016		1.7
Blank 1 Soil	organic	3.06	8.86	2490	91	1000	0.11	0.01	0.048	0.005		26
Blank 1 Soil	residual	3.06	16.7	1574	83	500	0.29	0.03	0.13	0.01	0.18	71
Pond C2 #3	exchangeable	3.00	16.7	4861	7	1000	0.01	0.003	0.004	0.001		0.74
Pond C2 #3	carbonate	3.00	16.7	4033	33	1000	0.05	0.01	0.021	0.004		4.2
Pond C2 #3	sesquioxide	3.00	16.7	4847	77	1000	0.09	0.01	0.040	0.005		8.1
Pond C2 #3	organic	3.00	16.8	5520	25	1000	0.03	0.005	0.011	0.002		2.3
Pond C2 #3	residual	3.00	16.7	1984	329	500	0.92	0.05	0.42	0.02	0.49	85
Pond C2 #5	exchangeable	3.00	16.7	4947	7	1000	0.01	0.003	0.004	0.001		0.15
Pond C2 #5	carbonate	3.00	16.7	4029	26	1000	0.04	0.01	0.016	0.003		0.68
Pond C2 #5	sesquioxide	3.00	16.7	5578	268	1000	0.27	0.02	0.12	0.01		5.0
Pond C2 #5	organic	3.00	16.8	5688	345	1000	0.34	0.02	0.15	0.01		6.4
Pond C2 #5	residual	3.00	16.7	1979	1652	500	4.6	0.2	2.1	0.1	2.4	88
SID 25	exchangeable	3.03	16.7	4294	4	1000	0.01	0.003	0.002	0.001		0.12
SID 25	carbonate	3.03	16.7	3890	18	1000	0.03	0.01	0.011	0.003		0.59
SID 25	sesquioxide	3.03	16.7	5319	90	1000	0.09	0.01	0.042	0.004		2.2
SID 25	organic	3.03	16.8	5238	402	1000	0.43	0.02	0.19	0.01		9.9
SID 25	residual	3.03	16.7	211	144	500	3.8	0.4	1.7	0.2	1.9	87
SID 27	exchangeable	3.02	16.7	4961	17	1000	0.02	0.005	0.0085	0.0021		0.39
SID 27	carbonate	3.02	16.7	4353	14	1000	0.02	0.005	0.0080	0.0021		0.36
SID 27	sesquioxide	3.02	16.7	5591	158	1000	0.16	0.01	0.070	0.006		3.2
SID 27	organic	3.02	16.8	5941	638	1000	0.60	0.02	0.27	0.01		12
SID 27	residual	3.02	16.7	58	43	500	4.1	0.8	1.8	0.4	2.2	84

35

Sample	Fraction	Mass (g)	Pu-242 (dpm)	Tracer Counts	Pu-239/240 Counts	Count Time (m)	Sample Activity (dpm/g)	Counting Uncertainty (1σ)(dpm/g)	Sample Activity (pCi/g)	Counting Uncertainty (1σ)(pCi/g)	Σ(Activity) (pCi/g)	Percent of Total (%)
SID 39	exchangeable	3.01	16.7	3062	8	1000	0.014	0.005	0.0065	0.0023		5.38
SID 39	carbonate	3.01	16.7	3580	8	1000	0.012	0.004	0.0056	0.0020		4.60
SID 39	sesquioxide	3.01	16.7	5661	14	1000	0.014	0.004	0.0062	0.0017		5.09
SID 39	organic	3.01	16.8	5078	12	1000	0.013	0.004	0.0059	0.0017		4.89
SID 39	residual	3.01	33.53	1703	33	500	0.22	0.038	0.097	0.017	0.12	80.05
Blank 2 soil	exchangeable	3.02	16.7	4264	10	1000	0.01	0.0041	0.006	0.002		7.5
Blank 2 soil	carbonate	3.02	16.7	3676	14	1000	0.02	0.0056	0.009	0.003		12
Blank 2 soil	sesquioxide	3.02	16.7	5579	18	1000	0.02	0.0042	0.008	0.002		10
Blank 2 soil	organic	3.02	16.8	3889	11	1000	0.02	0.0048	0.007	0.002		9.1
Blank 2 soil	residual	3.02	16.7	105	2	500	0.11	0.0752	0.047	0.034	0.078	61

Variable evaluated in duplicates:

Soil sample #2: variation in extraction time (min).	Exchangeable fraction:	2	65	2D	1170
	Carbonate:	2	310	2D	1080
	Sesquioxide	2	370	2D	655
	Ogranic	2	70 & 180	2D	305 & 240
	Residual	2	na	2d	na

Soil sample #3 and 4: KNO_3 *versus* $MgCl_2$

36

Appendix 6: Carbon Analysis.

Analysis Summary Microsoft Excel File: REFTS Carbon 9-97

Carbon Analysis: Rocky Flats Soil Samples
Analyst: R. Harnish
Analysis Dates: 9-13, 9-19, and 9-20-97
Method: Coulometrics titration; Total Carbon and Inorganic Carbon determined directly, in duplicate; Organic Carbon calculated by difference between TC and IC.

Sample	Total Carbon (% by weight)	Inorganic Carbon (% by weight)	Organic Carbon (% by weight)
#97L1879-001	2.56	0.17	2.39
#97L1879-002	2.32	0.12	2.20
#97L1879-003	2.41	0.77	1.64
#97L1879-004	2.35	1.36	0.99
#97L1879-005	2.23	0.95	1.28
#97L1879-006	2.33	0.16	2.17
Soil Blank from back of Coolbaugh	0.38	0.21	0.17
SID # 25	3.71	0.04	3.67
SID # 27	5.16	0.13	5.03
SID # 39	2.47	0.40	2.07
Pond C-2 # 3	0.74	0.25	0.49
Pond C-2 # 5	2.64	0.53	2.11
REFTS Soil C2-02	0.95	0.15	0.80

Total Carbon Analysis: Rocky Flats Soil Samples
Analyst: R. Harnish
Analysis dates: 9-13-97 and 9-19-97
Method: Coulometric titration; duplicate samples

Sample	Analysis 1 Sample Weight (mg)	Analysis 1 µg Carbon	Analysis 1 Percent Carbon	Analysis 2 Sample Weight (mg)	Analysis 2 µg Carbon	Analysis 2 Percent Carbon	Mean Percent Carbon	Mean deviation	Percent Mean Deviation
#97L1879-001	13	342.5	2.64	26	645.1	2.48	2.56	0.08	3.10%
#97L1879-002	50	1227	2.45	70	1526.2	2.18	2.32	0.135	5.80%
#97L1879-003	33	886.4	2.69	45	959.3	2.13	2.41	0.28	11.62%
#97L1879-004	33	813.4	2.46	23	516.2	2.24	2.35	0.11	4.68%
#97L1879-005	34	726.5	2.14	41	949.2	2.31	2.23	0.085	3.82%
#97L1879-006	59	1353.5	2.29	33	782	2.37	2.33	0.04	1.70%
Soil Blank	70	261.8	0.37	61	232.1	0.38	0.38	0.005	1.33%
SID # 25	15	565.8	3.77	21	766.2	3.65	3.71	0.06	1.62%
SID # 27	64	3460	5.41	19	932.1	4.91	5.16	0.25	4.84%
SID # 39	44	1070.3	2.43	39	979.7	2.51	2.47	0.04	1.62%
Pond C-2 # 3	67	498.3	0.74	63	466.9	0.74	0.74	0	0
Pond C-2 # 5	56	1577.8	2.82	37	910	2.46	2.64	0.18	6.82%
REFTS Soil C2-02	54	507.5	0.94	33	316.3	0.96	0.95	0.01	1.05%

Standards		Measured µg C	Theoretical µg C	Error
$CaCO_3$	10 mg	1226.2	1200	2.17%
$CaCO_3$	12 mg	1467	1440	1.90%
$CaCO_3$	8 mg	942.7	960	-1.80%
$CaCO_3$	13 mg	1527.9	1560	-2.10%

38

Inorganic Carbon Analysis: Rocky Flats Soil Samples

Analyst: R. Harnish
Analysis date: 9-20-97
Method: Coulometric titration; duplicate samples

Sample	Analysis 1			Analysis 2			Mean Percent Carbon
	Sample Weight (mg)	µg Carbon	Percent Carbon	Sample Weight (mg)	µg Carbon	Percent Carbon	
#97L1879-001	82	135.6	0.16	61	108.8	0.18	0.17
#97L1879-002	78	89.5	0.11	83	93.0	0.12	0.12
#97L1879-003	101	792.6	0.78	71	530.6	0.75	0.77
#97L1879-004	123	1661.9	1.35	112	1538.1	1.37	1.36
#97L1879-005	70	671.8	0.96	74	689.3	0.93	0.95
#97L1879-006	70	109.5	0.16	63	98.0	0.16	0.16
Soil Blank	175	422.4	0.20	116	243.9	0.21	0.21
SID # 25	59	22.6	0.03	78	36.4	0.05	0.04
SID # 27	65	80.2	0.12	69	88.4	0.13	0.13
SID # 39	101	380.8	0.37	45	189.1	0.42	0.40
Pond C-2 # 3	50	124.7	0.25	72	182.6	0.25	0.25
Pond C-2 # 5	127	651.8	0.51	101	553.9	0.55	0.53
REFTS Soil C2-02	166	172.1	0.10	145	167.7	0.20	0.15

Standards		Measured µg C	Theoretical µg C	Error
$CaCO_3$	10 mg	1222.3	1200	1.86%
$CaCO_3$	15 mg	1830.7	1800	1.71%
$CaCO_3$	9 mg	1102.4	1080	2.07%

Appendix 7. Summary of Pond Core Homogenates.

Sample	Dates Counted	Weight (g)	Wet wt.-W (g)	Wet wt.-A (g)	Dry wt.-A (g)	θ [--]	h (cm)	^{239,240}Pu (dpm/kg)	Error	^{239,240}Pu (Σdpm)	$\Sigma^{239,240}$Pu (Σdpm/cm^2)	$\Sigma^{239,240}$Pu (ΣpCi/cm^2)
97-05-29												
B501A	07/07-07/13	1.0057	183.62	19.641	12.5163	0.587	20	205.7	20.8	24.07	4.27	1.94
B502A	07/07-07/13	1.0085	160.26	15.998	9.9291	0.604	20	359.8	29.1	35.79	7.13	3.24
B503A	07/09-07-11	1.0063	163.28	14.705	9.0815	0.608	17.5	757.7	48.1	76.40	13.0	5.91
B504A	07/09-07/13	1.0058	171.52	19.646	12.4848	0.589	17.5	466.0	26.1	50.79	8.38	3.81
B506A	07/11-07/13	1.0095	174.67	18.205	11.3608	0.601	20	1266.7	67.0	138.07	25.28	11.49
B506A	08/20-08/26	1.0043	174.67	18.205	11.3608	0.601	20	490.8	55.0	53.49	9.79	4.45
B507A	07/13-07/18	1.0032	211.50	19.364	14.1781	0.478	30	178.0	13.5	27.57	6.97	3.17
B501B	07/18-07/21	1.0012	188.93	21.037	13.8556	0.564	20	219.5	18.9	27.32	4.80	2.18
B502B	07/18-07/21	1.0037	169.64	11.492	6.8784	0.626	20	446.9	31.5	45.37	8.36	3.8
B503B	07/21-07/23	1.0077	216.23	15.613	10.4213	0.555	17.5	672.6	42.3	97.07	13.11	5.96
B504B	07/21-07/27	1.0005	182.21	15.760	9.182	0.642	17.5	252.5	15.8	26.81	3.96	1.8
B506B	07/21-07/23	1.0092	186.39	21.739	13.372	0.610	20	372.1	28.6	42.66	7.26	3.30
B507B	07/21-07/27	1.0064	214.68	19.599	14.7761	0.449	30	91.3	6.8	14.78	3.76	1.71
B507B	08/11-08/17	1.0022	214.68	19.599	14.7761	0.449	30	148.3	15.6	24.01	6.14	2.79

40

Sample	Dates Counted	Weight (g)	Wet wt.-W (g)	Wet wt.-A (g)	Dry wt.-A (g)	θ [--]	h (cm)	$^{239,240}Pu$ (dpm/Kg)	Error	$^{239,240}Pu$ (Σdpm)	$\Sigma^{239,240}Pu$ (Σdpm/cm^2)	$\Sigma^{239,240}Pu$ (ΣpCi/cm^2)
97-05-20												
C202A	06/30-07/07	1.003	188.30	13.835	9.3948	0.542	10	182.6	12.3	23.35	2.09	0.95
C203A	06/30-07/07	1.0035	201.97	18.555	12.5882	0.542	12.5	416.6	25.2	57.08	5.96	2.71
C204A	06/30-07/07	1.0046	137.38	31.991	23.7736	0.464	10	138.8	11.2	14.17	1.85	0.84
C205A	06/30-07/07	1.0034	134.81	24.121	12.8724	0.686	10	3983.2	74.1	286.56	27.5	12.5
C205A	08/11-08/17	1.0014	134.81	24.121	12.8724	0.686	10	5984.9	334.7	430.56	41.36	18.8
C206A	07/07-07/09	1.003	111.94	19.334	12.7867	0.561	10	4585.3	141.6	339.45	50.31	22.87
C207A	07/07-07/09	1.0055	105.42	30.264	18.6641	0.608	10	5846.8	218.3	380.13	57.33	26.06
C202B	07/13-07/18	1.0019	147.38	24.028	15.6032	0.574	10	159.4	17.3	15.25	1.69	0.77
C203B	07/13-07/17	1.0021	123.36	11.980	8.7433	0.481	10	703.4	61.0	63.33	11.4	5.18
C203B	08/11-08/17	1.0044	123.36	11.980	8.7433	0.481	12.5	780.9	61.8	70.31	12.67	5.76
C204B	07/13-07/17	1.0022	196.09	23.011	16.2	0.512	12.5	275.7	27.4	38.06	3.37	1.53
C205B	07/17-07/21	1.0026	116.05	19.675	9.4044	0.732	10	4912.9	155.1	272.53	32.91	14.96
C206B	07/17-07/21	1.0031	181.67	16.146	10.8881	0.547	10	5205.2	262.8	637.68	58.96	26.8
C207B	07/18-07/21	1.0024	121.61	18.086	10.8583	0.625	10	5855.1	193.3	427.49	25.74	11.7

41

Sample	Dates Counted	Weight (g)	Wet wt.-W (g)	Wet wt.-A (g)	Dry wt.-A (g)	θ [--]	h (cm)	$^{239,240}Pu$ (dpm/Kg)	Error	$^{239,240}Pu$ (Σdpm)	$\Sigma^{239,240}Pu$ (Σdpm/cm^2)	$\Sigma^{239,240}Pu$ (ΣpCi/cm^2)
97-06-24-B1												
1	08/18-08/20	1.0053	107.79	14.0848	6.8038	0.728	35	2.44E+05	6653.5	12678.56	5.81×10^3	2.64×10^3
2	08/20-08/21	1.0043	131.95	15.162	10.5719	0.520	35	7.57E+04	2669.0	6967.52	3.19×10^3	1.45×10^3
4	08/18-08/20	1.0012	94.592	12.5494	5.7474	0.747	35	1.60E+05	4890.1	6912.35	3.61×10^3	1.64×10^3
5	08/20-08/21	1.0043	111.08	15.9793	7.4548	0.741	35	2.16E+05	8309.1	11210.71	4.91×10^3	2.23×10^3
6	08/18-08/20	1.0026	148.17	15.1248	8.7998	0.642	35	1.69E+05	2719.4	14565.87	5.3×10^3	2.41×10^3
7	08/18-08/20	1.0047	102.60	12.9456	5.5991	0.766	30-	1.12E+05	2880.9	4975.54	1.96×10^3	0.89×10^3

42

APPENDIX 8: CORE B5-5 ANALYSIS.

Sample	Dates Counted	Weight (g)	θ [–]	$^{239,240}Pu$ (dpm/kg)	Error	$^{239,240}Pu$ (Σdpm/cm^2)	$^{239,240}Pu$ (ΣpCi/cm^2)
(0-1 cm)	07/23-07/27	1.0031	0.772	1298.2	95.7	0.74	3.37E-01
(2-3 cm)	08/03-08/08	1.0043	0.663	259.0	26.0	0.22	9.91E-02
(2-3 cm)	09/05-09/11	1.0014	0.663	399.2	26.8	0.34	1.53E-01
(4-5 cm)	07/24-07/27	1.0071	0.679	3282.6	159.2	2.64	1.20E+00
(7-8 cm)	08/03-08/06	1.0021	0.624	756.6	44.3	0.71	3.23E-01
(9-10 cm)	07/23-07/27	1.0042	0.658	598.6	35.5	0.51	2.32E-01
(9-10 cm)	08/11-08/17	1.0064	0.658	1701.1	67.4	1.45	6.60E-01
(12-13 cm)	08/04-08/06	1.0017	0.635	1401.0	83.2	1.28	5.81E-01
(14-15 cm)	07/24-07/27	1.0054	0.612	878.1	59.9	0.85	3.87E-01
(17-18 cm)	08/04-08/08	1.0017	0.590	646.2	53.0	0.66	3.01E-01
(19-20 cm)	07/25-07/29	1.0085	0.621	1226.2	66.9	1.16	5.28E-01
(22-23 cm)	08/08-08/10	1.0006	0.624	2133.0	136.3	2.01	9.12E-01
(24-25 cm)	07/27-07/31	1.0037	0.626	1022.8	58.7	0.96	4.34E-01
(27-28cm)	08/06-08/10	1.0048	0.516	320.3	28.4	0.39	1.76E-01
(29-30 cm)	07/27-08/01	1.0036	0.444	128.9	18.5	0.18	8.15E-02
(32-33 cm)	08/06-08/10	1.0027	0.420	116.9	12.9	0.17	7.71E-02
(34-35 cm)	07/31-08/03	1.001	0.357	67.4	14.9	0.11	4.92E-02
(37-38 cm)	08/08-08/11	1.0027	0.365	33.5	11.2	0.05	2.42E-02
(39-40 cm)	08/01-08/03	1.0098	0.369	6.6	4.6	0.01	4.71E-03
(42-43 cm)	08/08-08/11	1.0081	0.385	84.6	22.2	0.13	5.91E-02
(44-45 cm)	08/03-08/08	1.0094	0.383	117.9	19.6	0.18	8.26E-02
(44-45 cm)	09/05-09/11	1.0033	0.383	40.4	6.6	0.06	2.83E-02
(46-47 cm)	08/01-08/03	1.0065	0.402	8.2	5.8	0.01	5.61E-03

Appendix 9: CORE C2-1 ANALYSIS.

Sample	Dates Counted	Weight (g)	θ [−]	^{239,240}Pu (dpm/kg)	Error	^{239,240}Pu (Σdpm/cm^2)	^{239,240}Pu (ΣpCi/cm^2)
(0-1cm)	07/21-07/23	1.0054	0.672	4797.2	188.2	3.93	1.79
(2-3cm)	07/21-07/23	1.002	0.633	4607.2	260.5	4.23	1.92
(4-5cm)	07/21-07/23	1.0064	0.567	1941.5	105.0	2.10	0.95
(7-8cm)	08/09-08/11	1.0081	0.515	2945.0	181.2	3.57	1.62
(9-10cm)	07/23-07/24	1.0044	0.650	2839.6	158.1	2.48	1.13
(12-13cm)	08/10-08/17	1.0031	0.512	4490.2	138.4	5.48	2.49
(14-15cm)	07/23-07/24	1.0049	0.650	4621.1	217.0	4.04	1.84
(14-15cm)	08/11-08/17	1.0054	0.650	5795.4	164.1	5.07	2.30
(17-18cm)	08/10-08/17	1.0061	0.527	1825.0	77.1	2.16	0.98
(18-19cm)	07/23-07/25	1.0068	0.512	1275.4	74.4	1.56	0.71

Appendix 10. $^{239,240}Pu$ K_d Values for 903 Pad 'lip' Area Soils.

Sample	Dates Counted	Weight (g)	$^{239,240}Pu$ (dpm/kg)	Error (1σ)
97L1879				
1	09/04-09/05	1.0021	36155.9	1357.1
1	09/07-09/08	1.0048	34879.6	653.3
2	09/04-09/05	1.008	68378.8	1652.6
3	09/04-09/05	1.0074	30947.8	1335.2
4	09/04-09/05	1.0024	19056.5	883.6
5	09/04-09/07	1.0038	4046.6	123.0
6	09/05-09/07	1.0044	49959.3	1222.7

Sample	Dates Counted	(ml)	(dpm/ml)	Error	Weight Sed. (g)	Kd ($cm^3\ g^{-1}$)	Error (1σ)	pH initial	pH after 5 days
1 & 491	09/15-09/21	500	3.69	0.09	5.0467	$0.98*10^4$	$\pm 0.04*10^4$	7.98	8.39
2 & 491	10/06-10/08	500	5.91	0.19	5.0041	$1.16*10^4$	$\pm 0.04*10^4$	7.89	8.3
3 & 491	10/06-10/08	500	0.86	0.07	5.0032	$3.59*10^4$	$\pm 0.33*10^4$	7.89	8.4
4 & 491		500	0.22	0.03	5.0087	$8.7*10^4$	$\pm 1.3*10^4$	7.96	8.39
5 & 491		500	0.03	0.01	5.002	$1.35*10^5$	$\pm 0.45*10^5$	7.98	8.21
6 & 1786	09/15-09/21	500	0.43	0.03	5.0427	$1.16*10^5$	$\pm 0.09*10^5$	7.49	7.94

Appendix 11. Determination of U(VI)/solar Pond core K_d Values.

Core 54294

Depth in core	Mass core sample (g)	Nitrate	pH	Fraction U(VI) sorbed [–]	Fraction U(VI) in solution [–]	Kd (L/kg)	Average Kd	Std. Dev. (1 σ)
top	10	high	7.324	0.6052	0.3948	153.3		
	10	high	7.426	0.5933	0.4067	145.9		
	10	high	7.304	0.5948	0.4052	146.8	148.7	4.0
	10	low	7.127	0.6338	0.3662	173.1		
	10	low	7.119	0.6325	0.3675	172.1		
	10	low	7.241	0.6267	0.3733	167.9	171.0	2.8
middle	10	high	7.436	0.3575	0.6425	55.6		
	10	high	7.426	0.3636	0.6364	57.1		
	10	high	7.414	0.3527	0.6473	54.5	55.8	1.3
	10	low	7.304	0.3679	0.6321	58.2		
	10	low		0.3752	0.6248	60.1		
	10	low		0.3695	0.6305	58.6	59.0	1.0
bottom	10	high	7.564	0.2417	0.7583	31.9		
	10	high		0.2274	0.7726	29.4		
	10	high		0.2447	0.7553	32.4	31.2	1.6
	10	low	7.345	0.3117	0.6883	45.3		
	10	low		0.3073	0.6927	44.4		
	10	low		0.3819	0.6181	61.8	50.5	9.8

Core 41193

Depth in core	Mass core sample (g)	Nitrate	pH	Fraction U(VI) sorbed [--]	Fraction U(VI) in solution [--]	K_d (L/kg)	Average K_d	Std. Dev. (1 σ)
top	10	high	7.584	0.5277	0.4723	111.7		
		high		0.5079	0.4921	103.2		
		high		0.5232	0.4768	109.7	108.2	4.5
		low	7.433	0.5422	0.4578	118.4		
		low		0.5522	0.4478	123.3		
		low		0.562	0.438	128.3	123.4	4.9
middle		high	7.604	0.3349	0.6651	50.4		
		high		0.3046	0.6954	43.8		
		high		0.3407	0.6593	51.7	48.6	4.2
		low	7.396	0.376	0.624	60.3		
		low		0.3516	0.6484	54.2		
		low		0.3384	0.6616	51.1	55.2	4.6
bottom		high	7.36	0.3578	0.6422	55.7		
		high		0.355	0.645	55.0		
		high		0.3841	0.6159	62.4	57.7	4.0
		low	7.323	0.3903	0.6097	64.0		
		low		0.4018	0.5982	67.2		
		low		0.3362	0.6638	50.6	60.6	8.8

APPENDIX 12: BOREHOLE LOGS FOR SOLAR POND CORES 41193 AND 54294.

3 Pages Following.

U.S. DEPARTMENT OF ERGY ROCKY FLATS PLANT

FORM GT.1A (REV. 2)

ROCKY FLATS PLANT BOREHOLE LOG

PAGE 1 OF 2

Borehole Number: 41193
Location - North: 751044 East: 2084873
Date: Drilled: 1-28-93 Logged: 8-20-93
Geologist: Reg Geo: C. Murray; Geo Logger: J. Evans
Drilling Equip.: CME 75, F900

Surface Elevation: 5960.70
Area: PA - NE of Pond 207A
Total Depth: 13.6'
Company: AE/RUST
Project No.: 40104
Sample Type: Dry Core

EG&G LOGGING SUPERVISOR APPROVAL C Hodge

DATE 8-24-93

TOP/BOTTOM OF CORE IN BOX	TOP/BOTTOM OF INTERVAL	FEET OF CORE IN INTERVAL (FIELD MEASUREMENT)	SAMPLE NUMBER	FRACTURE ANGLE	BEDDING ANGLE	GRAIN SIZE DISTRIBUTION	USCS SYMBOL	DEPTH IN FEET	SOIL/LITHOLOGIC LOG	SAMPLE DESCRIPTION
Box 1 of 2	0.0 Run 1 1.0	0.5	(1.35-1.60')			21G 36S 30SL 13C	SM			SM: Silty SAND with some gravel and clay (0.0-0.3'). Light brownish gray (10YR 6/2) to brown (10YR 4/3). Sand f.g. to c.g., well graded. Gravel f.g., low plasticity fines with abundant plant roots and organic matter from 0-0.1'. 2% Gravel, 36% Sand, 30% Silt, 13% Clay
	Run 2 2.0	0.8	V			67G 13S 18SL 2C	GM	1		
	Run 3 3.0	0.6	BH40050AE					2	NR	GM: Silty GRAVEL with some ~~silt~~ sand and trace clay. (0.3-2.6') Matrix dark brown (10YR 3/3). Gravel c.g., up to 7cm in length, poorly graded, sub-rounded; Sand same as above, moist. 67% Gravel, 13% Sand, 18% Silt, 2% Clay
	Run 4 4.0	0.7	BH40051AE (5.5-5.75') BH40049AE (0-5.9')				GW/gm	3	NR	
	Run 5 5.0	0.6				74G 18S 6SL 2C		4	NR	GW/gm: Sandy GRAVEL with some silt and trace clay (3.0-7.8'). Matrix brownish yellow (10YR 6/6), Gravel pred. c.g., well graded, sub-rounded to angular (drilling fractured, pred gray quartz, some metamorphic rock fragments; sand well sorted, trace (5%) caliche, moist, 74% Gravel, 18% Sand, 6% Silt, 2% Clay.
	Run 6 6.0	0.9	V C					5	No recovery	
	R7 6.6	0.6						6	NR	
	Run 8 7.4	0.5						7	NR	
	Run 9 7.8	0.4	BH40053AE (9.75-10.1') BH40052AE (6-7.9') C							Top bedrock at 7.8
	Run 10 9.0	0.8			0°	8S 70SL 22CL		8	NR	Clayey SILTSTONE with some sand (7.8-11.0') Light gray (10YR 7/1). Slightly friable, crude horizontal bedding, Sand v.f.g. to f.g. With scattered Mg-oxide staining trace Fe-oxide staining, moist, 8% Sand, 70% Silt, 22% Clay
	Run 11	1.3	V					9		
								10		

NOTES: General: USCS is modified for this log as follows:
Materials amounts are estimated by % volume instead of % weight.
(1) Badly broken core, accurate footage measurements not possible.
(2) Core breaks cannot be matched, accurate footage measurements ...

49

U.S. DEPARTMENT OF ERGY ROCKY FLATS PLANT FORM GT.1A (REV. 2)

ROCKY FLATS PLANT BOREHOLE LOG

PAGE ___ OF ___

Borehole Number: 41193
Location - North: 751044 East: 2034873
Date: Drilled: 1-28-93 Logged: 2-20-93
Geologist: R.G.C. Murray Logged: J. Evans
Drilling Equip.: CME 75, F901
Surface Elevation: 5160.70
Area: PA NE of Pond 207A
Total Depth: 13.6'
Company: AE/RVST Project No.: ___
Sample Type: Dry Core

EG&G LOGGING SUPERVISOR
APPROVAL C Hodel DATE 5-24-93

TOP/BOTTOM OF CORE IN BOX	TOP/BOTTOM OF INTERVAL	FEET OF CORE IN INTERVAL (FIELD MEASUREMENT)	SAMPLE NUMBER	FRACTURE ANGLE	BEDDING ANGLE	GRAIN SIZE DISTRIBUTION	USCS SYMBOL	DEPTH IN FEET	SOIL/LITHOLOGIC LOG	SAMPLE DESCRIPTION
	10.3									
Box 2 of 2	Rm12	2.2						11		Clayey SILTSTONE with some sand (11-12'). Yellow (10YR7/6) Slightly to moderately friable, with discontinuous thin clay laminations., moist 21% Sand, 47% Silt, 32% Clay
	12.5							12		
	Rm13	1.1						13		Clayey Siltstone with some sand, same as above (reference interval 7.8'-11.0'). With chaotic bedding possibly due to soft sediment deformation.
	13.6							14		Total depth drilled 13.6'

NOTES: General USCS is modified for this log as follows:
Materials amounts are estimated by % volume instead of % weight.
(1) Badly broken core, accurate footage measurements not possible.
(2) Core breaks cannot be matched, accurate footage measurements not possible.

50

U.S. DEPARTMENT OF ENERGY ROCKY FLATS PLANT

FORM GT.1A (REV. 2)

ROCKY FLATS PLANT BOREHOLE LOG

PAGE 1 OF 1

Borehole Number: 54294
Location - North: 751051.6 East: 2085006.5
Date: 8-9-94
Geologist: J.P. O'BRIEN
Drilling Equip.: HSA / CME 75

Surface Elevation: 5960.7
Area: Solar Ponds
Total Depth: 10.0
Company: ES
Project No.: OU4 GEOTECH
Sample Type: CONTIN. CORE / DRIVE

EG&G LOGGING SUPERVISOR APPROVAL: A. M. Pant

DATE: 11/22/94

TOP/BOTTOM OF CORE IN BOX	TOP/BOTTOM OF INTERVAL	FEET OF CORE IN INTERVAL (FIELD MEASUREMENT)	SAMPLE NUMBER	FRACTURE ANGLE	BEDDING ANGLE	GRAIN SIZE DISTRIBUTION	USCS SYMBOL	DEPTH IN FEET	SOIL/LITHOLOGIC LOG	SAMPLE DESCRIPTION
								1		
	2.0							2		START OF CORE @ 2.0'
	RUN 1	1.4				61% GRAVEL 33% SAND 5% SILT 1% CLAY	GW	3		SANDY GRAVEL - LIGHT GREY (10YR 7/2), MAX CLAST DIAMETER 3.5", AVE 1", FINE TO COARSE GRAINED SAND, WELL GRADED, GRAVEL IS SUB-ROUNDED TO FRACTURED, SAND IS ANGULAR TO SUBROUNDED, LOW PLASTICITY. GRAVEL IS CALICHE COATED QUARTZITE, SAND CONTAINS ABUNDANT QUARTZ AND ROCK FRAGMENTS WITH TRACE MICA, CALICHE HORIZON 3.15'-3.3', DRY. TOP OF BEDROCK @ 3.3'
								.3	.4	CLAYSTONE - GREENISH GREY (5GY 6/1) TO BROWNISH YELLOW (10YR 6/2) WHERE IRONSTAINED (6.4'-6.9', 7.4', 9.0'-9.2'), VERY FINE GRAINED, VERY WELL SORTED, ROUNDED TO WELL ROUNDED, ~4% POROSITY, ARGILLACEOUS CEMENT, NON-FRIABLE, ABUNDANT QUARTZ, CALICHE HORIZONS 3.3'-3.4', 5.3'-5.4', 6.6'-6.8', 7.5', THINLY TO THICKLY LAMINATED 7.6'-8.0', 9.0'-9.8', OTHERWISE MOTTLED, IRON STAINED 45° FRACTURE @ 7.4', IRONSTONE NODULES 9.0-9.2', MOIST.
	4.0							4		
	RUN 2	2.0	4.3 BH COARSED 5.1			5% SAND 10% SILT 85% CLAY		5		
	6.0							6		
Box 1 of 1	RUN 3	2.0	6.9 BH COARSED 7.6	45°				7		
	8.0							8		
	RUN 4	1.8						9		
	10.0							.8		T.D. @ 10.0'

Parsons Engineering Science, Inc.
QA Record No: _____ Page 25 of 38

QA Checked By: LRD Date 2/23/95

NOTES: General: USCS is modified for this log as follows:
Materials amounts are estimated by % volume instead of % weight.
(1) Badly broken core, accurate footage measurements not possible.
(2) Core breaks cannot be matched, accurate footage measurements not possible.

51/51